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MAGNETIC CORRELATIONS IN $La_{2-x}Sr_xCuO_4$ FROM NQR RELAXATION AND SPECIFIC HEAT

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ABSTRACT

We review ^{139}La and ^{63}Cu NQR spin-lattice relaxation rates and specific heat measurements as a function of temperature in $La_{2-x}Sr_xCuO_4$ for x ranging from 0 up to 0.3, with particular emphasis on the effect of doping on the Cu^{2+} spins magnetic correlations. In the low doping limit, $x \leq 0.05$, the results can be interpreted consistently in terms of a simple phenomenological "two-fluids" model whereby the effect of thermally-activated mobile O(2p) holes is the one of disrupting locally the Cu^{2+} spin correlations. For $x \geq 0.1$, the results indicate the onset, as $T \to T_c^+$, of a strong coupling between Cu^{2+} spins and the Fermi liquid of O(2p) holes leading to the apparent disappearance of localized Cu^{2+} moment in connection with the opening of a superconducting gap.

INTRODUCTION

A central issue for the understanding of the microscopic mechanism leading to superconductivity in Cu-base high-T_c superconductors is about the interplay between the O(2p) holes controlling the transport and the effective Cu magnetic moments controlling the magnetic properties. In both $La_{20x}Sr_xCuO_4$(LaSCO) and $YBa_2Cu_3O_{6+\delta}$(YBCO) systems one can change, as a function of composition, from a planar Heisenberg system of localized Cu^{2+} moments into a metallic system in which a strong coupling exists between the Cu^{2+} d-states and the itinerant O-p states. The true nature of the Cu^{2+} magnetic state and of its correlations in the latter case is still a matter of debate.

NMR-NQR and relaxation measurements have provided enlightening information about the above-mentioned problematic, particularly in the two systems YBCO and LaSCO. The studies of spin dynamics in YBCO by $^{63,65}Cu$ NMR-NQR have been recently reviewed.[1] Furthermore, ^{89}Y[2,3] as well as ^{17}O NMR[4-6] have proved to be sensitive tools to test the properties of Fermi liquid of O(2p) holes.

In this paper, we present a review of our ^{139}La and ^{63}Cu NQR relaxation measurements in LaSCO.[7] The results are analyzed in terms of a simple phenomenological model which leads to a satisfactory classification of the magnetic correlation properties of the Cu^{2+} d-states as a function of Sr-doping and as a function of temperature.

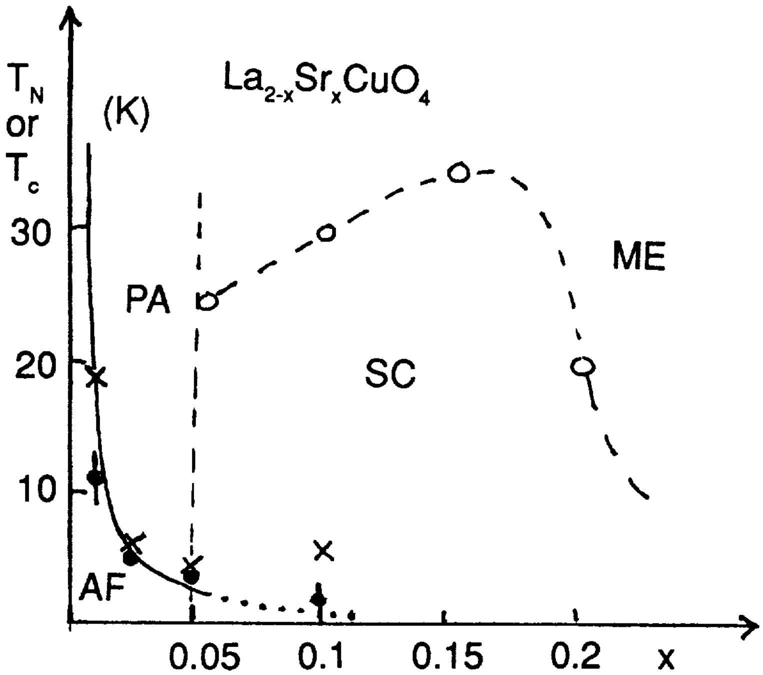

FIG. 1 Phase diagram for LaSCO based on results of the present investigation (see Ref. 7): (O) Meissner effect; (x) specific heat; (●) NQR. The solid line is the theoretical result, Eq. (12), in the text.

The phase diagram of LaSCO is shown in Fig. 1. For sake of simplicity, only our data referring to the samples which were used in the present investigation are indicated. The boundary between the paramagnetic (PA) and antiferromagnetic (AF) phase is obtained from NQR[7] and specific heat[8] measurements. The normal metal (ME)-superconductivity (SC) boundary is obtained from Meissner effect measurements.[7] The boundary between semiconducting and metallic phase is not defined by experimental measurements; in the subsequent discussion, we will assume it to be around $x = 0.05$. The boundary between tetragonal and orthorombic structure is not indicated since it is not relevant for the purpose of our investigation. It should be noted that there exist substantial agreement in the literature about the phase diagram of LaSCO except that for two points:

(i) the magnetically ordered phase observed at low temperature for $x \geq 0.01$ can be either a spin-glass phase or a disordered antiferromagnetic phase.

(ii) a coexistence of magnetic order and superconductivity has been reported for $x \geq 0.05$. Although we also find this coexistence in both NQR and specific heat measurements, we tend to interpret it as due to phase separation.

In the analysis of NQR T_1 data in LaSCO, it is useful to consider separately two ranges of x-doping:

(i) moderate doping, namely $x \leq 0.05$ where the samples are semiconductors and a transition from the paramagnetic to an ordered phase is observed.

(ii) strong doping, namely $x \geq 0.1$ or metallic region, where at T_c one has the transition to the superconducting phase.

NQR Relaxation and Spin Dynamics

The ^{139}La NQR spin-lattice relaxation rates are obtained by monitoring the growth of the magnetization due to the population difference between the $\pm 5/2 \leftrightarrow \pm 7/2$ levels, (frequency separation in the range $18 \div 19$ MHz) after a saturating sequence of rf pulses. The same holds for the ^{63}Cu nucleus in terms of the $\pm 1/2 \leftrightarrow \pm 3/2$ NQR levels (frequency separation in the range $35 \div 38$ MHz). The recovery process can be expressed in terms of relaxation transition probabilities W which can be due, in principle, to two mechanisms:

(i) time dependence of the electric field gradient (EFG) components induced by lattice vibrations and/or by defect motion (quadrupole mechanism)

(ii) fluctuations of the local hyperfine field h(t) acting on the nucleus and due to localized and/or itinerant electron spins (magnetic mechanism).

It was found[7] that the dominant mechanism is the magnetic one except, perhaps, for ^{139}La around room temperature.

The dominant magnetic relaxation transition probability driven by the fluctuations induced by the Cu^{2+} electronic spins $\vec{S}(t)$ on the hyperfine field $\vec{h}(t) = h_{eff}\ \vec{S}(t)$ at the nucleus (^{63}Cu or ^{139}La) is:

$$W_M = \frac{1}{4}\gamma_N^2 \int < h_+(0)h_-(t) > \exp(-i\omega_R t)dt \tag{1}$$

where ω_R is the resonance frequency for a given pair of NQR levels and the z axis is along the maximum component of the EFG tensor in its principal axis frame of reference. By introducing collective spin components $\vec{S}_{\vec{q}}$, in the assumption of isotropic spin fluctuations, and by taking into account that ω_R is much smaller than the characteristic frequencies of the spin fluctuations, the spectral density in Eq. (1) can be rewritten:[9]

$$J(\omega_R \simeq 0) = (h^0_{eff})^2 \frac{1}{N}\int \sum_q < S^z_q(0)S^z_{-q}(t) > dt \tag{2}$$

where h^0_{eff} represents the hyperfine field at the nucleus for the rigid-lattice AF structure of localized Cu^{2+} magnetic moments (namely for $T \to 0$ in the prototype system La_2CuO_4). The result in Eq. (2) is valid only if the hyperfine field is weakly $\vec{q}$ and $\alpha(\pm,z)$ dependent. This condition is fulfilled quite well for ^{63}Cu and approximately at the ^{139}La site.[7c]

By considering the relationship[7c] between W_M and the experimentally measured spin-lattice rates T_1^{-1}, one derives from Eqs. (1) and (2):

$$T_1^{-1} = \frac{1}{N} A \sum_q \int < S^z_q(0)S^2_{-q}(t) > dt = \frac{1}{N} A \sum_q \frac{|S_{\vec{q}}|^2}{\Gamma_q} \tag{3}$$

where we have assumed that the isotropic collective spin fluctuations decay exponentially with decay rate Γ_q. Here, $A_{La} = 11.5\ \gamma^2_{La}\left(h^0_{eff}\right)^2_{La}$ and $A_{Cu} = 3\gamma^2_{Cu}$. From the splitting of the NQR line below T_N, one obtains[7a,b] $\left(h^0_{eff}\right)_{La} \simeq 0.1$ Tesla(T) while from the ^{63}Cu zero field–NMR in La_2CuO_4 at 1.3 K one has[10] $\left(h^0_{eff}\right)_{Cu} = 7.8$ T, thus yielding

$$A_{La} = 1.6 10^{14} \qquad rad^2 s^{-2} \tag{4a}$$

$$A_{Cu} = 9.10^{17} \qquad rad^2 s^{-2} \tag{4b}$$

In normal exchange-coupled insulating magnetic systems, the relaxation rate T_1^{-1} can be related to the magnetic correlation length $\xi = k^{-1}$ in the framework of static and dynamic scaling:[9]

$$|S_{\bar{q}}|^2 = k^{-2+\eta} f_2(q/k) \tag{5a}$$

$$\Gamma_{\bar{q}} = Bk^z f_3(q/k) \simeq \omega_e k^z f_3(q/k) \tag{5b}$$

where z and η are characteristic critical exponents.[9] In the assumption $f_2 \cdot f_3 \simeq 1$ and $\eta = 0$ one has:

$$T_1^{-1} = A \frac{k^{\,d-2}}{\Gamma_c} \tag{6}$$

where $\Gamma_c = \omega_e k^z$ is the frequency corresponding to the critical fluctuations and d is the lattice dimensionality. In the limit of no correlation, i.e., T >> J/k_B where J is the Heisenberg exchange interaction one has:

$$\Gamma_c \simeq \omega_e = \left[\frac{8}{3}\frac{J^2 S(S+1)}{\hbar^2}\right]^{1/2} \tag{7}$$

NQR in the Low-Doping Semiconducting Phase

In the region of moderate doping ($x \leq 0.05$) it was found[7c,d] that the ^{139}La T_1^{-1} data are inconsistent with the predictions of Eq. (6) if one assumes that the correlation length is limited by thermal fluctuations, i.e. $\xi, \propto (T - T_N)^{-\nu}$. On the other hand, the ^{139}La relaxation data can be explained[7c,d] consistently if one assumes that the correlation length is limited by the presence of the mobile 0(2p) holes. This corresponds to the assumption that the local time-dependent correlation function of the Cu^{2+} spins decays exponentially to zero because of "collisions" with mobile defects rather than because of thermal fluctuations.

Then one can write, starting from Eq. (3):

$$T_1^{-1} = A \frac{\tau_d}{\Gamma_c \tau_d + 1} \simeq A\tau_d = A\tau_0 n^{-1} \tag{8}$$

where we introduce a "collision" time $\tau_d = \tau_0 n^{-1}$ inversely proportional to the effective concentration n of mobile defects. The assumption $\Gamma_c \tau_d << 1$ in Eq. (8) reflects the fact that the magnetic correlation is limited by the defects rather than by thermal fluctuations.

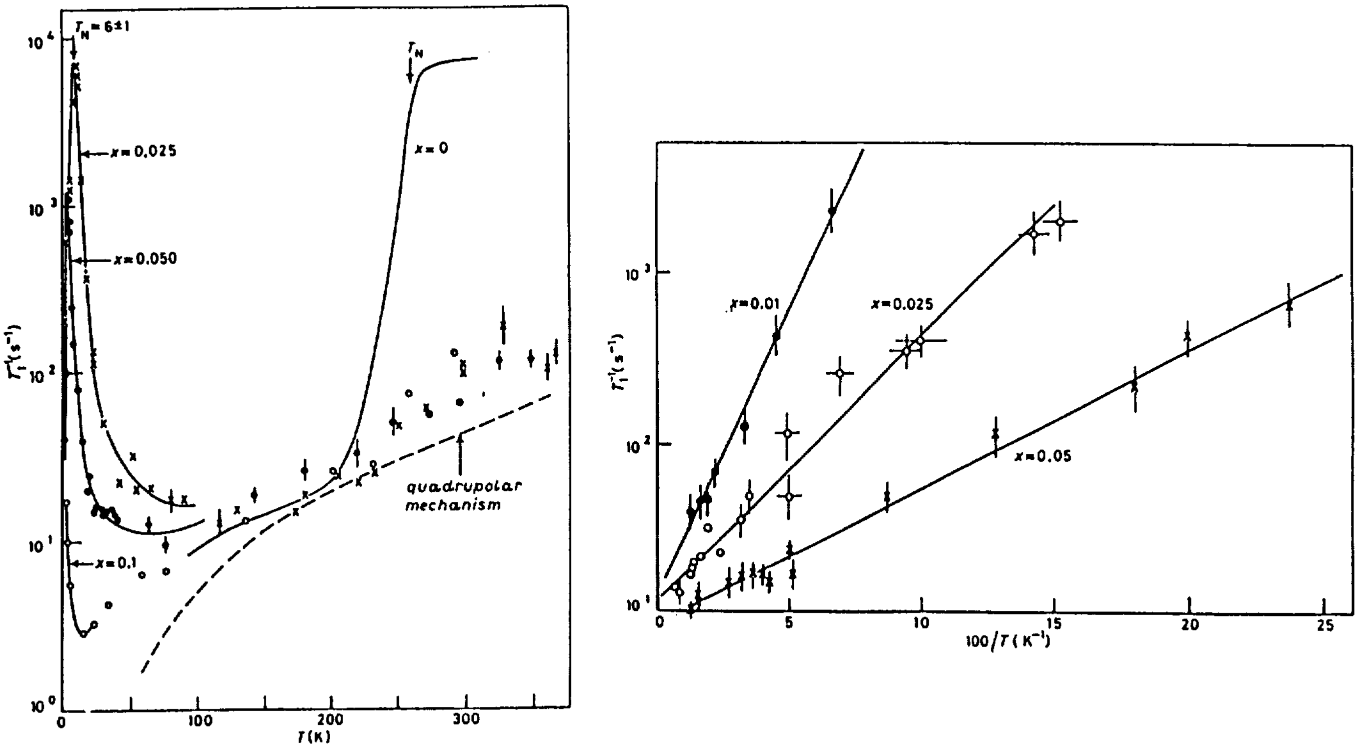

FIG. 2 ^{139}La NQR spin-lattice relaxation rates vs. temperature in $La_{2-x}CuO_4$. (a) The full lines are guides to the eye. The dashed line is the quadrupole contribution to relaxation as estimated theoretically in Ref. 7c. The position of the peak (T_1^{-1} vs. T) corresponds to the temperature T_N at which long range order sets in as reported in Fig. 1. (b) Semilog plot of T_1^{-1} vs T^{-1} used to obtain the activated law for the concentration n of mobile charge defects (see Eq. (9).)

By fitting Eq. (8) to the ^{139}La T_1^{-1} data for $x < 0.1$ (shown in Fig. 2), it was deduced[7c,d] that the concentration of mobile defects should follow a thermally-activated law

$$n(x,T) = x\exp\left(-0.85/xT\right) \tag{9}$$

with an x-dependent activation energy $E(x) = 0.85/x(^\circ K)$. This implies that as the temperature is lowered well below $\sim 0.85/x(K)$ the mobile defects "freeze" and the magnetic correlation length is no longer limited by the defects thus allowing the system to order at T_N (see Fig. 1). The fit of the ^{139}La T_1^{-1} yields $\tau_0 = 2.10^{-15}$ sec rad^{-1} which corresponds to the inverse of the exchange frequency, ω_e, defined in Eq. (7) for an exchange interaction J = 2700 K.

The present model leads to precise predictions[7c,d] regarding the x and T dependence of the correlation length ξ and of the ordering temperature T_N. Regarding ξ, one can note that the correlation length can be thought as the average distance among defects, i.e., $\xi \propto n^{-(1/d)}$ where d is the lattice dimensionality. Thus from Eq. (8), one has

$$T_1^{-1} = A\tau_0\xi^2 \tag{10}$$

where A is given in Eq. (4), $\tau_0 \simeq \omega_e$ as defined in Eq. (7) and we have taken d = 2 for the CuO_2 planes. From the experimental value for $^{139}La\ T_1^{-1}$ (see Fig. 2), one derives for ξ the values shown in Fig. 3. The theoretical curve:

$$\xi = \frac{1}{\sqrt{x}} \exp\left(\frac{0.85}{2xT}\right) \tag{11}$$

follows from our model and the result in Eq. (9). Regarding the x-dependence of T_N, it appears that the magnetic order sets in when the correlation length reaches a certain critical value ξ^* independent of concentration x. Setting $\xi^* = 100$ lattice units in Eq. (11) and solving for T, one has

$$T_N(x) = \frac{0.85}{2x\ell n(100\sqrt{x})} \tag{12}$$

The theoretical curve Eq. (12) is compared with the experimental values in Fig. 1.

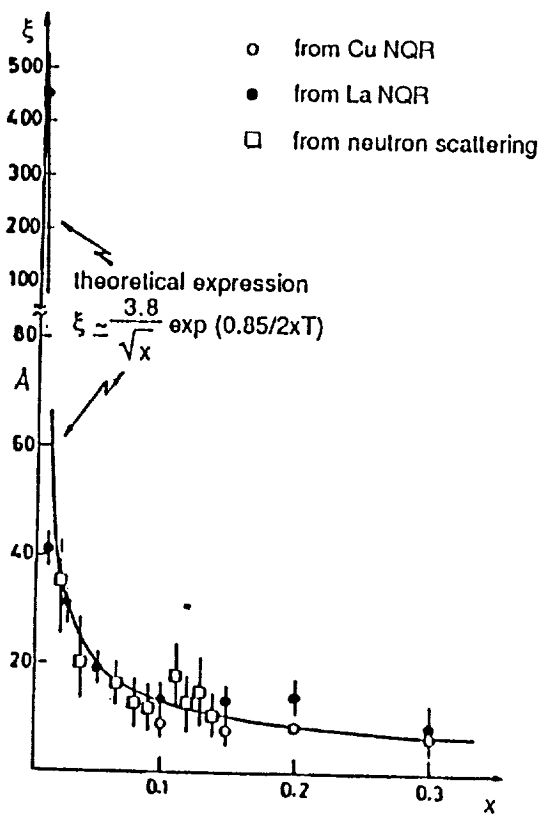

FIG. 3 In-plane magnetic correlation in $La_{2-x}Sr_xCuO_4$ at T = 77 K, in Å, as derived from NQR T_1 for ^{139}La and ^{63}Cu, according to Eq. (10) (the value for nominally pure La_2CuO_4 at $T \geq T_N$ is also reported) and comparison with the results obtained through neutron scattering (R. J. Birgeneau et al. *Phys. Rev. B* **39**, 2868 (1989).

It should be noted that in the analysis outlined above, the amplitude of the hyperfine magnetic field at the nuclear site has been assumed x and T independent and given by h^0_{eff} (see Eq. 4). Therefore, the role of the mobile 0(2p) holes for $x \leq 0.05$ is only the one of disrupting locally the strong AF correlations, without affecting the intrinsic magnetic moment of the Cu^{2+} ions. This corresponds to setting in Eq. (3) $< S_z^2 >= \frac{1}{N} \sum_q < |S_q|^2 > = 1$.

NQR in the High-Doping Metallic and Superconducting Phases

The NQR relaxation rates T_1^{-1} for both ^{63}Cu and ^{139}La in samples with $x \geq 0.1$ are characterized by a weakly temperature-dependent region at $T > 100$ K followed by a rapid decrease as the temperature is lowered through the superconducting critical temperature T_c (see Figs. 4 and 5). In order to analyze the data in these large doping metallic samples, one should refer to expressions of T_1^{-1} in terms of an x and T dependent generalized susceptibility $\chi(q,\omega)$ describing the coupled system of Cu 3d and O-2p holes.[11-13] However, since a rigorous treatment of this complex system is not yet available, we chose to interpret the data in terms of our simple phenomenological model which still retains the main physical insights. Starting from the "two fluid" model which appear to be valid in the low concentration semiconducting limit, i.e., $x \leq 0.05$, we write:

$$T_1^{-1} = A < S_z^2 > \tau_d + A_0^2 \chi_s^2(x,T)T \tag{13}$$

where we neglect the weak q-dependence of the coupling parameter A for both ^{63}Cu and ^{139}La nuclei. Furthermore, the hyperfine interaction A should not change appreciably as a function of temperature as indicated qualitatively by the almost constant value of the quadrupole coupling frequency ν_Q, which, just like A, depends on the symmetry and probability density of the electronic wave function at the nuclear site.[14] The second term in Eq. (13) is the Korringa-type contribution[14] related to the hyperfine interaction, A_0, of ^{63}Cu (or ^{139}La) with the 0-2p Fermi liquid described by the spin susceptibility χ_S.

(a) ^{63}Cu Spin-Lattice Relaxation Rate

The ^{63}Cu NQR relaxation rates are shown in Fig. 4 for x = 0.15 and x = 0.2. The solid line in Fig. 4 is the theoretical behavior expected for x = 0.2 by using for T_1^{-1} the expressions (8) and (9) corresponding to $< S_z^2 > = 1$. With the value of A_{Cu} in Eq. (4b) and $\tau_0 = 2.10^{-15}$ s, one has $T_1^{-1} = 1.8\ 10^3/x$ in satisfactory agreement with the experimental results at high temperature.

As the temperature is lowered, one has a dramatic difference between the samples with low x and the ones at large doping. For $x \leq 0.05$, the relaxation rate becomes so short that the ^{63}Cu NQR signal is undetectable, conforming to the notion that the Cu^{2+} spins retain their localized character, i.e., $< S_z^2 > = 1$. On the contrary, for $x \geq 0.1$, one observes a rapid decrease of T_1^{-1} as $T \rightarrow T_c^+$, opposite to the behavior expected from the solid line in Fig. 4 indicating that the "two fluids" model breaks down in this limit.

The decrease of T_1^{-1} on cooling can be described phenomenologically as due to a decrease of the degree of localization of the Cu^{2+} spins associated with the coupling to the liquid of O-2p holes. Since the contribution to the Cu relaxation rate from the carriers, second term in Eq. (13), is negligible (see Fig. 4), one can derive the temperature dependence of $< S_z^2 > \sim \sum_q \chi(\bar{q},\omega = 0)$ from Eq. (13) and the data in Fig. 4. The results are shown in Fig. 5(a). From the semilog plot of $h_{eff}(x,T) = h_{eff}^0 < S_z^2 >^{1/2}$ vs T^{-1} shown in Fig. 5(b) the temperature dependence appears to be of the form exp $(-\Delta/T)$ with an activation energy which is correlated to T_c: in fact the ratio $2\Delta/T_c$ is close to 3.5 as one would expect for conventional BCS mechanism with gap 2Δ.[15]

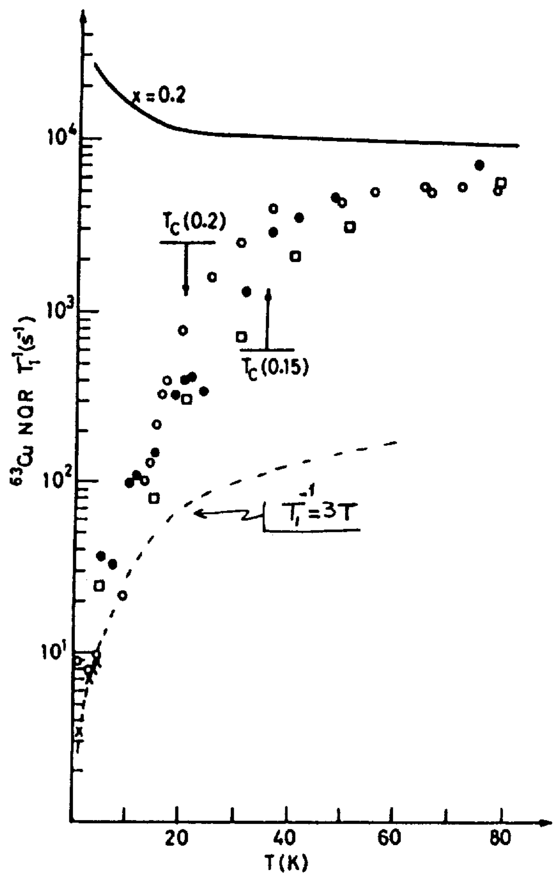

FIG. 4 ^{63}Cu NQR spin-lattice relaxation rates $T_1^{-1} = 6\ W_M$ vs. temperature in $La_{2-x}Sr_xCuO_4$ for: (O) x = 0.2; (●) x = 0.15. A few data from K. Ishida et al., *J. Phys. Soc. Jpn.* **58**, 36 (1989) are reported for comparison (□). The dashed line represents the Korringa-like contribution extrapolated from low temperature data (x) in x = 0.4 (Ref. 21). The solid line is the behavior expected from Eqs. (8) and (9) for a fully localized moment, i.e., $< S_z^2 > = 1$.

It should be emphasized that the physical interpretations of the decrease of $< S_z^2 >$ remains an open question. One could speculate that the apparent disappearance of the Cu^{2+} local moment could be just a way to describe the "quenching" of the local moment susceptibility due to the strong coupling with the conduction charges, a problem reminiscent of the Kondo effect found when a magnetic impurity is dissolved in a normal metal.

(b) ^{139}La Spin-Lattice Relaxation Rate

The ^{139}La NQR relaxation rates are shown in Fig. 6 for x = 0.2. The quadrupole contribution to the relaxation rate can be neglected in this temperature range[7] (see Fig. 2a). Therefore, one can use Eq. (13) to describe the two dominant relaxation mechanisms for ^{139}La. By using the results by Kobayashi et al.[16], for $x \geq 0.3$, one estimates for the ^{139}La Korringa term in Eq. (13), $A_0^2\chi_s^2 = 4.10^{-2}\ s^{-1}K^{-1}$ (a renormalization by a factor 10 is taken into account in this estimate because of a different definition of T_1 in terms of W_M used in Ref. 16). Two theoretical curves have been drawn in Fig. 6 to be compared with the experimental data. The first one (upper curve) is obtained by using Eq. (13) with the $< S_z^2 >$ extracted from the results for ^{63}Cu (see Fig. 5(a)), A from Eq. 4(a), $\tau_d = 10^{-14}x/(s.rad^{-1})$, and $A_0^2\chi_s^2 = 4.10^{-2}s^{-1}K^{-1}$ as given above. The second curve (lower curve in Fig. 6) is obtained as before, except for the scaling of the spin susceptibility in Eq. (13), according to $\chi_s(x,T) = \chi_s^0 < S_z^2 >^{1/2}$. This assumption takes into account the coupling of the Cu^{2+} spin with the O2p holes within the framework of our simple description. Because of the large uncertainty in the experimental results, it is not possible to decide which of the two descriptions is more appropriate. However, in either case, the temperature dependence of ^{139}La NQR relaxation for large x reflects unambiguously the temperature dependence of the localized Cu^{2+} magnetic moment in the same way as for ^{63}Cu NQR T_1.

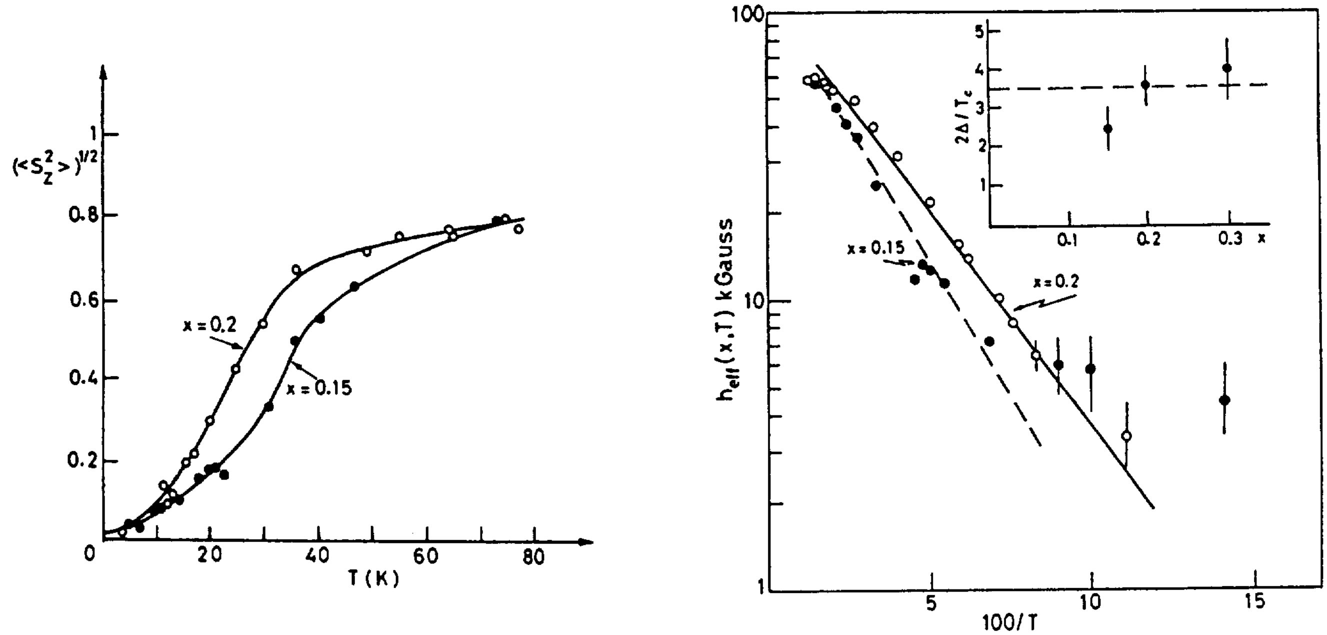

FIG. 5 (a) Temperature dependence of the normalized local Cu^{2+} spin moment, $(< S_z^2 >)^{1/2}$, in $La_{2-x}Sr_xCuO_4$ for x = 0.2 and x = 0.15 as obtained from the experimental data for ^{63}Cu NQR T_1 (Fig. 4) according to Eq. (13) in the text. Note that $T_c(0.2) = 20 \pm 5K$ and $T_c(0.15) = 35 \pm 5K$.

(b) Semilog plot of $h_{eff}(x,T) = h^0_{eff}(< S_z^2 >)^{1/2}$ vs. 100/T obtained by using the temperature-dependence of $< S_z^2 >^{1/2}$ shown in part (a) and $h^0_{eff} = 7.8$ T. The straight lines correspond to activation energies Δ (0.2) = 36 $\pm$ 3K and Δ (0.15) = 40 $\pm$ 10K. For x = 0.3 one obtains a more uncertain estimate of Δ around 10 K. In the inset, the ratio $2\Delta/T_c$ is shown as a function of x.

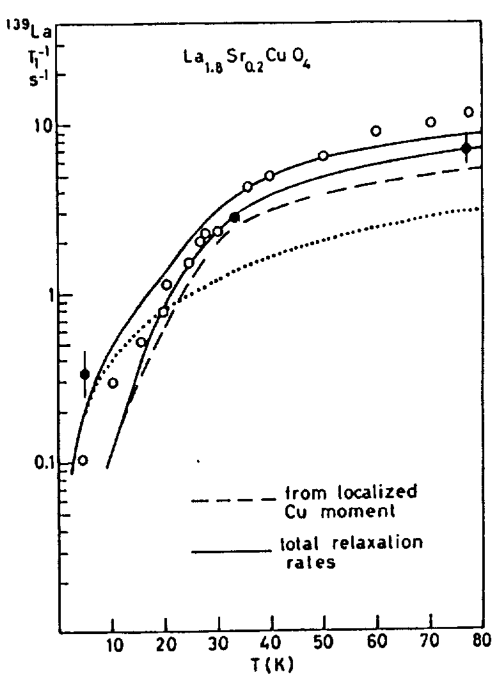

FIG. 6 Temperature dependence of ^{139}LA NQR relaxation rate in $La_{2-x}Sr_xCuO_4$ for x = 0.2. The experimental points are from Ref. 16 (O) with representative points measured by us (●). The dotted line represents the Korringa-like behavior obtained for x = 0.4 and x = 0.3 from Ref. 16 as explained in the text. The dashed line is the theoretical behavior according to the first term in Eq. (13) and using the temperature dependence of $< S_z^2 >$ shown in Fig. 5 (see text.). The two solid lines represent the total rates according to Eq. (13) as explained in the text.

Specific Heat Measurements

The samples with x = 0.01, 0.025, 0.05, and 0.1 show an anomaly in the specific heat which can be ascribed to the onset of long-range magnetic order[8] (see Fig. 7). The corresponding transition temperatures, T_N, derived from the position of the specific heat peak, are plotted in Fig. 1. As can be seen, our results compare well with the ones obtained from other techniques except for x = 0.1. The sharpness of the peak observed in the specific heat is more in favor of a 3D antiferromagnetic transition, as in the prototype La_2CuO_4, rather than a spin-glass type transition. It should be stressed that the anomalies reported here were not observed in the previous measurements, although a specific search for them does not seem to have been done.[17]

At high and intermediate temperatures, the effect of substituting La with Sr should be related mostly to the changes in the phonon spectrum.[8] For T < 15 K and for x = 0.05 and x = 0.025, there seems to be a hint of an x dependent broad maximum superimposed to the background specific heat. This feature could be associated with the excitation spectrum of the O-2p holes and/or the effect of the holes on the Cu^{2+} spin magnetic correlations. The contribution to the specific heat can be estimated in the framework of the heuristic model proposed to explain the NQR results. In such a model, it is hypothesized that the electronic holes are trapped at low temperature and become mobile with an activation energy E_a which is inversely proportional to x. By assuming that the mobile holes behave as a classical gas of noninteracting particles of concentration $\bar{n}$(x,T) one has:

$$\frac{C_v}{k_B} = E_a\frac{\partial\bar{n}(x,T)}{\partial T} = \left(\frac{0.85}{T}\right)^2\frac{1}{x}\frac{\exp(-0.85/xT)}{(1+\exp(-0.85/xT))^2} \tag{14}$$

where we have used for $\bar{n}$(x,T) the normalized expression (9):

$$\bar{n}(x,T) = \frac{x\exp(-E_a/k_BT)}{1+\exp(-E_a/k_BT)} \tag{15}$$

with $E_a/k_BT = b/xT$ (b=0.85 K). The theoretical behavior of Eq. (14) is compared with the experimental data in Figs. 7a,b. Although the agreement is rather poor, one should keep in mind that Eq. (14) was derived on the basis of an oversimplified model and that the parameters found in the NQR analysis were used without any adjustment.

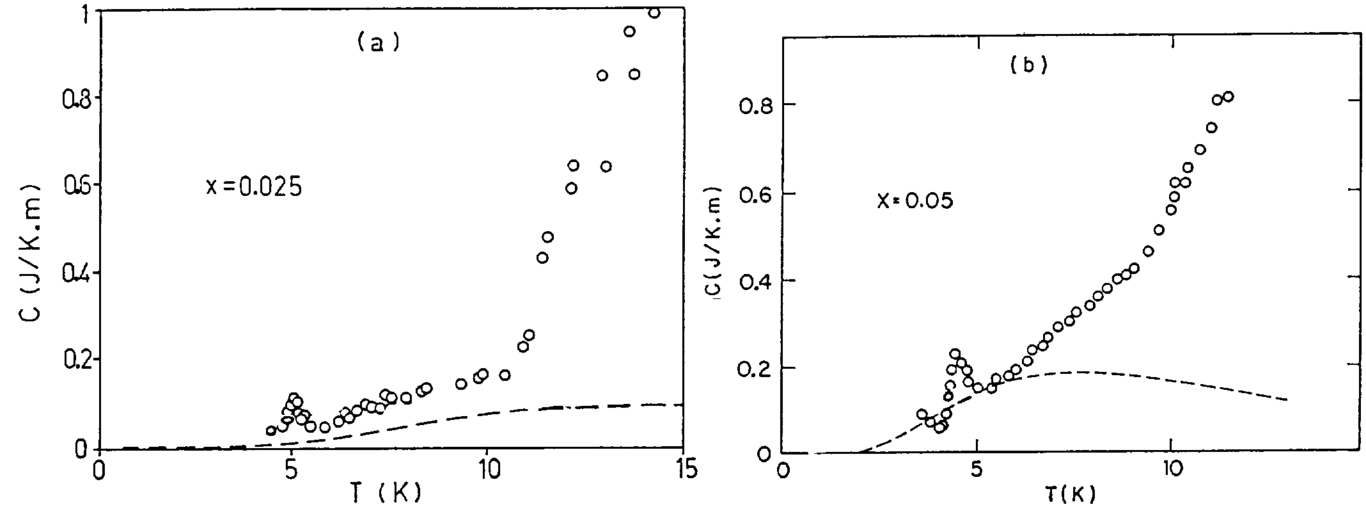

FIG. 7(a,b) Specific heat vs. temperature in $La_{2-x}Sr_xCuO_4$ for two Sr concentrations compared with the theoretical curve (Eq. (14) and (15) in the text) representing the contribution of the thermally excited O 2p mobile holes.

SUMMARIZING REMARKS

It has been shown that in the superconducting $La_{2-x}Sr_xCuO_4$ a decrease on cooling of the effective Cu^{2+} local spin moment occurs, possibly as a result of the pairing processes occurring in the liquid of charge carriers. This reduction, for large doping ($x > 0.1$) is observable well above T_c and it is consistent with susceptibility[18] as well as μSR[19] measurements. The temperature dependence of the effective Cu^{2+} spin appears to be well-described by an activated law, with a gap 2Δ correlated to T_c. If the decrease of $< S_z^2 >^{1/2}$ is indeed related to the opening of a gap, it is remarkable that the effect is observed well above T_c. Analogous "precursor" effects were detected in YBCO ($T_c = 61$ K) in the temperature range $90 \div 100$ K, and interpreted in terms of pair formation and energy gap beginning in the CuO_2 plane above the three-dimensional superconductivity.[1]

Numerical studies of the 2D Hubbard model have produced results for the dependence of the local spin moment from temperature and from the one-site repulsion U.[20] If analogous results could be obtained for the dependence of the Cu^{2+} local spin moment from the coupling to the thermally fluctuating Fermi liquid of O2p holes, they could be used for a more quantitative comparison with our experimental findings.

An important debated issue regards the possible coexistence of superconductivity and long-range magnetic order. Low temperature NQR spectra[21] for $x \geq 0.1$ were interpreted as indicating magnetic ordering, with reduced Cu^{2+} magnetic moments. However, more recent ^{63}Cu NMR-NQR spectra[22] in aligned polycrystalline samples seem to rule out the existence of a sizeable static hyperfine field at $T = 4.2$ K. The peak in the specific heat in the sample at $x = 0.1$ (see Fig. 1 and Ref. 8) could also be interpreted in terms of a phase separation or inhomogenity of the sample (the Meissner effect yielding $T_c = 28$ K for $x = 0.1$ is only partial, i.e., $\simeq 50\%$). Finally, no evidence of a static magnetic moment in $La_{1.85}Sr_{0.15}CuO_4$ at $T = 20$ mK was found in the time evolution of the muon polarization function.[23]

A comparison with recent findings by neutron scattering in LaSCO[24] and in YBCO[25] is in order. While T_1 is sensitive to the q-integrated generalized susceptibility in the MHz range, inelastic neutron scattering yield, in principle, $\chi''(q,\omega)$ in the whole frequency range, for a selected wave vector. However, a frequency resolution limit around 1 meV has to be taken into account, thus making difficult the comparison with the NQR relaxation rate. In particular, neutron scattering seems to indicate that around T_c the spin dynamics of the Cu^{2+} spins undergo a change: the normalized integrated intensity of the magnetic response function exhibits a drastic decrease in the low energy range, while it is not T-dependent for energies larger than about 15 meV, whereby the q-integrated spin is conserved.[25] Since no static hyperfine field at the Cu site can be detected below T_c, these modes would have to be interpreted as paramagnons in a strongly correlated itinerant antiferromagnetic system without long range order. In this case, the decrease of $< S_z^2 >^{1/2}$ observed by us (see Fig. 5) would have to be ascribed to a progressive changeover from a diffusive correlation function centered at zero frequency into an oscillatory correlation function having a power spectrum centered at ω_c, thus far above the NQR resonance frequency. The main conclusions regarding the temperature dependence of the effective hyperfine interaction due to the Cu^{2+} spins and of the spin susceptibility of the Fermi liquid, with the precursor effects above T_c are not modified.

We mention that the physical picture at the basis of the interpretative model outlined in this paper can be used consistently also to explain the ^{63}Cu, ^{89}Y, and ^{17}O NMR-NQR results in YBCO-type systems, as it is discussed elsewhere.[7c]

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